

not found in naturally occurring elemental silicon, exhibit properties unlike bulk silicon.

an Various techniques exist for producing silicon nanoparticles of random dimensions, and those processes sometimes produce silicon nanoparticles below 10 nm.--

Please replace the paragraph beginning on page 3, line 11, with the following rewritten paragraph:

--A discretely and uniformly sized family of silicon nanoparticles are provided by the method of the invention. Execution of the basic method steps includes a gradual advancing electrochemical etch of bulk silicon with low-intermediate current density. A density of ~ 10 milli Ampere per square centimeter ($10\text{mA}/\text{cm}^2$) is optimal to produce the family. Higher current density favors 1nm particles, and lower the larger particles. Blue (1nm) particles, if any appreciable quantity exist depending on the selected current density, may be removed by, for example, shaking or ultrasound. After the etch, the pulverized wafer is immersed in dilute HF for a short time, while the particles are still connected to the wafer, to weaken the linkages between the larger particles. This may be followed by separation of nanoparticles from the surface of the silicon. Once separated, various methods may be employed to form plural nanoparticles into crystals, films and other desirable forms. The nanoparticles may also be coated or doped. The invention produces the family of a discrete set of sized particles and not a continuous size distribution. The discrete sizes in the family of particles are 1 (blue emitting), 1.67 (green emitting), 2.15 (yellow emitting), 2.9 (red

emitting) and 3.7 nm (infrared emitting) nanometer nanoparticles. The silicon nanoparticles
a2 may also be combined or reconstituted into crystals, solids, films, etc.--

Please replace the paragraph beginning on page 4, line 4, with the following
rewritten paragraph:

--The method for creating the elemental silicon nanoparticle family of the invention is an electrochemical treatment which involves gradually advancing bulk silicon, e.g., a wafer, into a chemical etch bath at a low speed while etching is taking place, in the presence of an external current. A meniscus forms as a very thin slice of the silicon that is at the interface of etchant solution and air. The slow advance of the silicon creates a large section, which is meniscus-like. In effect, a traveling meniscus is created as the silicon material is gradually advanced into the etchant bath while electrical current is applied. At this point, any blue (1nm) particles may be removed by shaking, scraping, or, preferably, gentle
a3 ultrasound. After the etch (and any removal of blue particles), the pulverized wafer is immersed in dilute HF for a short time, e.g., about 1 minute, while the particles are still connected to the substrate to weaken the linkages between the larger particles. The overall process enriches the ultra small substructure of the material. Moreover, it makes the top skin of the silicon material extremely fragile. The ultra-small structures, which are silicon nanoparticles, may then be separated from the material and recovered. Our procedure, using the intermediate-low current densities and the dilute post HF immersion primarily produces

a3 the larger green, yellow, red and infrared nanoparticles of the present family. Further separation or filtering is effective in isolating particles of a given size with a high degree of uniformity.--

Please replace the paragraph beginning on page 5, line 9, with the following rewritten paragraph:

a4 --The current density will determine the sizes of particles formed. An optimal current density for forming the range of particles is ~ 10 milli Ampere per square centimeter ($10\text{mA}/\text{cm}^2$). At that level, though, a very small percentage of the blue (1nm) particles will form. Raising the current density produces more 1nm particles at the expense of larger particle production. For example, $20\text{ mA}/\text{cm}^2$ has been found to produce good results that favor production of 1nm particles. Reducing current density to $10\text{ mA}/\text{cm}^2$ produces the family of particles. Further reduction to $5\text{ mA}/\text{cm}^2$ further favors larger particles. If the goal is to produce only 1nm particles, then the current density is increased. At the higher current density, e.g., $20\text{ mA}/\text{cm}^2$, the most stable size is the 1 nm, the smallest stable size we have been abundantly producing. The procedure is straightforward and always leads to the monodispersed 1 nm nanoparticles. At the intermediate densities or low current densities, several larger sizes of particles (1.67, 2.15, 2.9, or 3.7 nm) result from the etching procedure, excluding (or including to a much lesser degree) the smallest 1 nm ones. The 1nm particles, if any, are very tenuously connected to the silicon substrate and other particles, however the

94 larger sizes have stronger linkages to each other and the substrate. Thus, following the etch and prior to the HF immersion it is possible to shake off those smallest ones (if any), e.g., by a gentle ultrasound, a shaking, or a scraping. At this point, the recovery method for the larger sizes diverges. After the etch, the pulverized wafer is immersed in dilute HF for a short time, e.g., about 1 minute, while the particles are still connected to the substrate to weaken the linkages between the larger particles. The wafer is transferred to the liquid of choice, and ultrasound is used to shake the particles off, allowing them to be dispersed into the solution. At this point the solution then contains a mixture of the family of sizes excluding the 1 nm size nanoparticles.--

Please replace the paragraph beginning on page 7, line 13, with the following rewritten paragraph:

95 --Once a colloid of the larger green, yellow, red and infrared particles is obtained, these larger particles may also be isolated from each other by additional processing. In a preferred method of isolation, we centrifuge the mix of particles. The residue consists of the largest particles (red and infrared) in the mix. The smaller green/yellow particles stay in suspension in the liquid. The residue of the centrifuge will then be recovered and re-dissolved in fresh solvent and sonificated (to separate red and infrared from the residue). The largest red emitting particles will stay in suspension, while the residue (infrared) is discarded. A harsher solvent treatment and ultrasound may be conducted to obtain the largest infrared

from the residue. The green particles may be isolated by additional sonification/or and the addition of a minute quantity (e.g., a drop of HF) acid with an ultrasound to break up the green from the yellow particles. The yellow particles are more difficult to separate from the green. *as* Chromatography is best to accurately separate the yellow and green particles. Accordingly, the initial isolation process may be followed by commercial Gel permeation chromatography to separate the yellow from the green ones further, if necessary, or to obtain additional accuracy in separation of the other particles, as well. The particles are separated into vials each containing particles of uniform size, with near 90-100 % efficiency.--

Please replace the paragraph beginning on page 8, line 3, with the following rewritten paragraph:

a6 --A commercial method of isolating particles is chromatography. The commercial Gel permeation chromatography is also known as size exclusion chromatography and is based on separation by size. There is no dependence on the molecular weight. The basic setup is a column that is packed with porous gel beads through which a solvent flows. The sample is injected into the solvent upstream of the column. The column is packed with gel beads, composed of a polymer, that have pores of various sizes on them. The smallest pores define the lower limit of sizes that the column can separate. Particles that are as small as or smaller than this lower limit have the largest fraction of pores available for them to diffuse into. The upper limit of size that can be separated is determined by the largest pore

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size in the beads. The gel does not trap anything larger. With a continuous distribution of pore sizes, separation is possible for all particles sizes between the upper and lower limits. The separation occurs due to the time delay in the particle's drift through the column caused by the particles being trapped in the pores. The larger the number of available pores, the slower the particles traverse the column, leading to a physical separation of particle sizes as a function of the time they exit the column resulting in a distribution of five discrete highly uniform size distributions. The mix is separated into vials each containing particles of uniform size with near 90-100 % efficiency.--

Please replace the paragraph beginning on page 8, line 25, with the following rewritten paragraph:

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--The silicon nanoparticles of the invention have good electronic, chemical and structural qualities. The preferred use of H_2O_2 as part of the etchant solution to produce the silicon nanoparticles provides a high quality hydrogen coating (termination or passivation), dominated by an ideal stretching mode with no di- or tri hydrides that act as nonradiative electronic traps. The high quality coating fully protects the silicon particles from uncontrollable low quality post interactions in the ambient atmosphere that would generate nonradiative traps. Moreover, the preferred etchant leaves no oxygen on the particles. After the electrochemical etching process is complete, the hydrogen coating, however, can be replaced by a high quality oxide coating by post immersion in H_2O_2 . This is due to the fact

a7 that the high quality nature of the hydrogen termination makes it possible for hydrogen to be substituted with an ultra thin high quality oxide termination. This is a self-limiting process that yields oxide layers of 4 Å thick on bulk Si, with a trap density of less than $1/10^{14}$. The oxide coating provides additional protection and hardening against laser damage. The optical properties of the silicon nanoparticles are not compromised by such a high quality oxide termination (passivation). As to the chemical quality, H_2O_2 is an oxidant which will remove nearly every kind of contamination (organic material, metals, alkalines, and metal hydroxides) from silicon surfaces by oxidative dissolution and complex formation.--

REMARKS

Applicants have amended the Specification to place it in better form grammatically. No new matter has been added.

Attached hereto is a marked-up version of the changes made to the specification and claims by the current amendment. The attached Appendix is captioned **“Version with markings to show changes made.”**